ARKIV FÖR FYSIK Band 3 nr 35

Communicated 6 June 1951 by AXEL E. LINDH and BENGT EDLÉN

On the concentration of heavy water in glacier ice

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With 2 figures in the text

As early as in 1934 EUCKEN and SCHÄFER (1) tried to determine the concentration of deuterium oxide in the water from glacier ice. Their results showed that glacier ice probably contains a higher per cent of deuterium oxide than does the ordinary subsoil water. Their investigations concerned specimens from some of the big Swiss glaciers. The following investigation was started to ascertain whether, likewise, a high concentration of heavy water is to be found in the comparatively small glaciers of the North Scandinavian mountain range.

Experimental Technique

In their investigations EUCKEN and SCHÄFER made use of a method of measuring densities first published in 1934 by MOSER (2). This method demands comparatively voluminous samples as well as elaborate temperature measurements. Moreover, the temperature of the thermostat has to be kept constant on 10^{-3} degrees and less. This seemed to be too elaborate a technique only for ascertaining, whether there is a surplus of heavy water in the glacier ice. Also, as the Swedish glaciers are to be found in regions with only primitive communications, it was desirable, for reasons of transport, to make use of a measuring technique needing only small samples. After some abortive experiments with floating bodies a method was employed, which was invented by LINDESTRØM-LANG, JACOBSEN and JOHANSEN (3). Here the sample itself constitutes the floating body, the liquid consisting of a mixture of oils. A tube shaped as A in Fig. 1 is placed in a thermostat. The lower bulb and the lower half of the tube between the two bulbs contains a mixture of bromobenzene and kerosene. This fluid has a density slightly higher than that expected for the samples. The upper bulb contains a mixture of the same sort of oils but with a density slightly lower than that of the samples. Beginning midway between the two bulbs the two fluids will start to mix, eventually exhibiting a continuous increase of density with the depth. Usually mechanical stirring is needed to start the mixing of the two fluids, the densities being nearly the same.

The test tube is evaluated by putting small drops of water with a known surplus of deuterium oxide down the tube. These drops will arrange themselves on different levels according to their density. A drop of the sample is

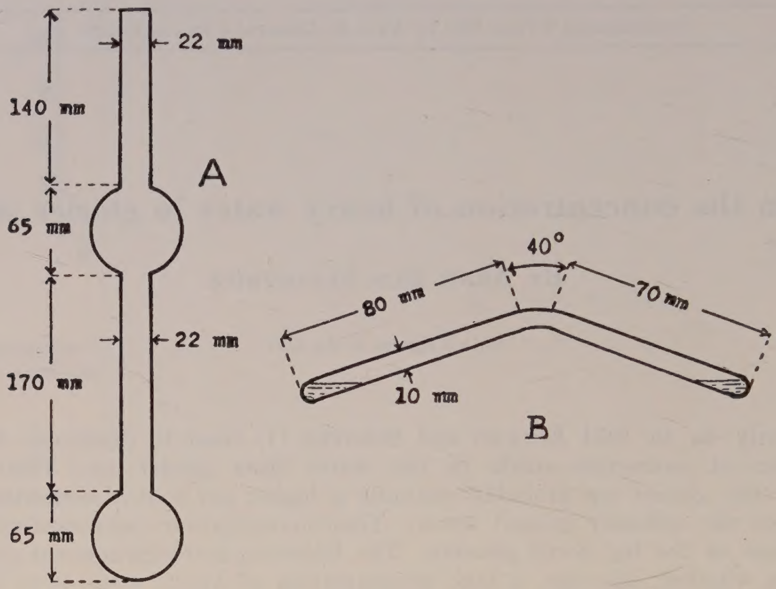

Fig. 1. The shape and the dimensions of the test tubes (A) and the pyrex tubes for the distillation process (B).

then inserted and its position in equilibrium is determined relative to the drops with known densities. With some skill it is possible to give all the drops the same size. To prevent water from the drops to dissolve in the oil mixture, the latter ought to be loaded with water either by the method described by LINDERSTRØM-LANG and coworkers (3) or by leaving the tube for some time with drops of water in contact with the oil mixture. Two or three tubes were always kept simultaneously ready for use.

Already at an early stage of the experiments it was found that owing to the small differences in density the method of LINDERSTRØM-LANG and coworkers had to be changed slightly. It turned out to be rather difficult to work with a long series of reference drops. Therefore, only two reference drops were used, one from common subsoil water and the other with a density higher than that of the sample. The distances between the three drops were measured with a cathetometer and the surplus of heavy water in the sample determined by linear interpolation. Naturally, by using only two reference drops, the accuracy is considerably lower than that of the original method.

It ought to be remarked here that some early measurements were made on photographs of the relative positions of the drops. It was found, however, that the precision attained did not justify the additional work.

Naturally, it is necessary to use as pure samples as possible. Glacier ice carries a lot of contaminations, the larger particles consisting mainly of dust from the surrounding rock. Most of this dust can be removed by filtering. A certain amount of sodium chloride is always present and sometimes also a considerable amount of iron oxide (4), most of which passes through the densest filters in use.

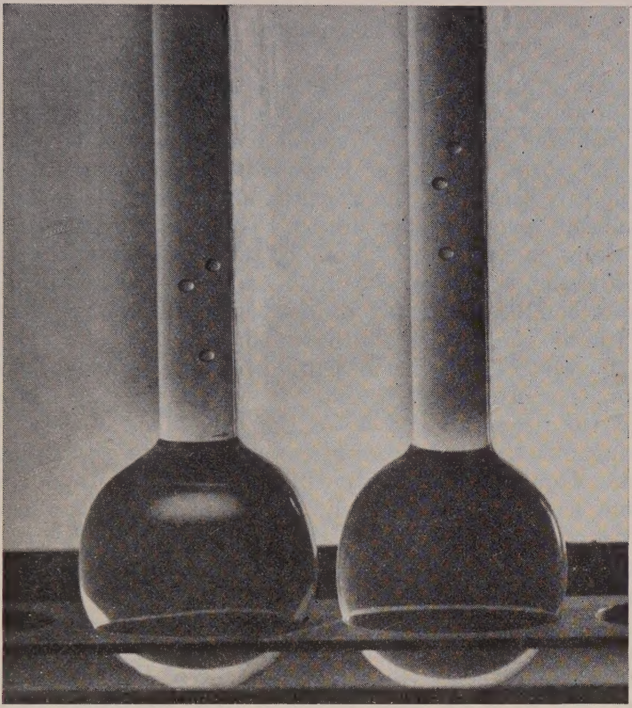

Fig. 2. Two test tubes with the water drops floating in the oil mixture. The density difference between the reference drops is $24.5 \cdot 10^{-6}$ units.

The samples were purified from most of the solid contaminations by being filtered as soon as they were brought to the laboratory. They were stored in small bottles closed by stop cocks until shortly before measurements were to start, when they were distilled according to LINDERSTRØM-LANG and co-workers (3).

A small portion of the sample was put into a pyrex tube shaped like tube B in Fig. 1. The part of the tube containing the water is placed in a cavity in a copper block, which is cautiously heated. The water condenses in the other end of the tube. When a sufficient amount has been collected, the tube is allowed to cool, after which the purified water is used to clean out the condenser part of the pyrex tube. This procedure is repeated at least twice. When at last the definitive distilling process is undertaken, great care is taken that, in the end, all the water is collected in the clean half of the tube. All the time a close watch has to be kept lest any of the contaminated water should sputter into the condensate. When the procedure is finished, the tube is blown off in the bend and the sample preserved in the closed tube until drops are to be brought out with a pipette. A more complete description of the procedure was given by LINDERSTRØM-LANG and co-workers (3).

The results

Fig. 2 is a photograph of two of the test tubes side by side in the thermostat. The small water drops are clearly visible, although their spherical form is somewhat distorted by the cylindrical lens action of the tubes with their oil mixture. The drops are rather large, having diameters of 2.5 mm. There is no doubt in this case that the density of the samples is higher than that of common subsoil water (upper drops). The density difference between the two reference drops is $24.5 \cdot 10^{-6}$ units. In this special case the smallest detectable density difference would be $2.5 \cdot 10^{-6}$ units but when the diffusion of the two oil mixtures had been effected in the best possible way, density differences less than 10^{-6} units could easily be detected.

Although a method of relative measurements was employed the temperature had to be kept as constant as possible. It was often very difficult to avoid a slow motion of the drops up and down the test tube, as even the light, necessary for carrying out the measurements, was sufficient to disturb the temperature conditions. However, the relative positions of the drops did not change, once equilibrium had been established. A certain difficulty was experienced in inserting the drops along the axis of the tube, a slight sidewise motion eventually causing the drops to touch the wall. As soon as this happened the drop adhered to the glass. This was one of the reasons why it was difficult to work with a series of more than two reference drops.

The samples were collected from several glaciers. A number of them are from the Kebnepakte glacier, which ends with a vertical ice wall. The samples I to VII and XX to XXV were collected from the lower part of this wall. They constitute two groups collected with an interval of one year. The results regarding their surplus of heavy water are collected in Table I. Numbers within parentheses refer to values which, in one way or another, are to be

Table I.

Samples from the ice wall of the Kebnepakte glacier.

Sample No.	Excess of heavy water in per cent of total sample	Average excess of heavy water
I	0.036	Samples I to VII 0.049
II	0.042	
III	0.052	
IV	0.029	
V	(0.066)	
VI	0.077	
VII	(0.031)	
XX	0.022 0.033 0.048 (0.030) 0.052	Samples XX to XXV 0.053
XXI	0.046 0.056 0.059	
XXII	(0.065) (0.168) (0.074) 0.099	
XXIII	(0.010) 0.073	
XXIV	(0.00) 0.041	
XXV	(0.025) (0.00)	

Table II.

Single samples from a number of glaciers.

Sample No.	Origin of sample	Excess of heavy water in per cent of total sample
VIII	The Kebnepakte glacier half-way up its course	0.027
XII	Small glacier on the summit of Kebnekaise, 2121 m	0.055 0.036
XV	Surface ice on the Björling glacier	Density the same as that of subsoil water
XVII	Crevasse, lower part of a glacier on Sjelmatjokko	0.109
XVIII	Crevasse, upper part of Storjökeln, Kebnekaise	0.048
101	Surface layer, upper part of the Aka glacier, Sarek National Park	0.038
103		0.033
104	Surface layer half-way up the Pårte glacier, Sarek National Park	0.046
107	Crevasse, Isfallsjökeln, Kebnekaise	0.050

considered as unreliable, the reason being, mostly, that one of the three drops was destroyed before a sufficiently long series of observations had been collected. These values were not included in the calculations of the averages.

The results regarding samples from other glaciers are collected in Table II. These include one sample from the Kebnepakte glacier collected half way up its course. Some measurements were also made on water from other sources than the massive glacier ice. The results are to be found in Table III.

As can be seen from sample XX the values vary considerably. In this case the four values refer to drops from four separate distillates. However, the values vary inside very wide limits even when the drops were taken from one and the same distillate. Accordingly, it is most unprobable that variations such as those in the case of sample XX arise from an inferior distillation technique. A rather obvious difference was observed, also, when drops from one and the same distillate were introduced into two of the test tubes placed side by side in the thermostate. This showed beyond doubt that the variations arise from the density of the test fluid not increasing linearly with the depth. When only two reference drops were used this variation could not be accounted for. Although part of the error can be eliminated by averaging, the limits of error will still be very wide.

Table III.

Water samples from the vicinity of glaciers.

Sample No.	Origin of sample	Excess of heavy water in per cent of total sample
XIV	Névé, Björling's glacier	0.022
IX	Brook from the last remnants of large snowfields	0.021
X	Water from Lake Tarfala	Density probably the same as that of subsoil water
XI	Snow from the upper part of Storjökeln, Kebnekaise	
XIII	Snow on the South Peak, Kebnekaise	
XXVI	Water falling down the ice wall of the Kebnepakte glacier.	
102	Snow from the upper part of the Aka glacier, Sarek National Park	

Discussion

The results show without any shadow of doubt that water from the ice of the Swedish glaciers has a density higher than that of ordinary subsoil water. As far as our knowledge goes it seems right to ascribe this increased density to an excess of heavy water. According to Table I and II this excess varies between 0.02 and 0.10 per cent corresponding to an enrichment of not less than one to five times the normal concentration of heavy water.

If the thirteen samples from the ice wall of the Kebnepakte glacier are considered as a group from the same homogeneous material, altogether thirty-six relatively reliable measurements are available for the calculation of a mean value. However, regardless of the wide limits of errors the distribution of values shows that the material is not to be regarded as homogeneous. This is certainly due to the fact that the content of heavy water varies considerably from one part of the bulk of ice to another. Such a variation was also observed by Eucken and Schäfer, and is to be expected from the manner in which the ice probably is enriched with heavy water. Accordingly, the mean values in Table I are to be taken as the average excess of deuterium oxide in the ice.

It is interesting to note that the average excess of heavy water in the ice from other glaciers is of the same order of magnitude as that of the ice from the Kebnepakte glacier (Table II). A sample from the small glacier on the summit of Kebnekaise is of special interest. This glacier consists of a thin ice cap covering the summit and part of the ridge. The zones of accumulation and ablation coincide. The sample was collected at the rim, where the ice-sheet is thinning down over the rocks below.

In their first paper EUCKEN and SCHÄFER (1) gave a thermodynamical theory of the enriching process. According to their notions the actual enrichment takes place during the process of melting. If this were true a considerable excess of heavy water is to be expected in the zone of ablation but not in the upper parts of a glacier. Some of the results presented in Table II are at variance with this. Excepting that from the Kebnekaise summit all the samples listed in this table were collected in parts of the glaciers far above the "tongue". According to EUCKEN and SCHÄFER the ice of the lower part of a glacier with a broad front ought to show a lower concentration of heavy water than the ice from the corresponding part of a glacier ending in a narrow tongue. Most of the Swedish glaciers are comparatively short and broad and thus rather low concentrations ought to be expected. Nevertheless, the results listed in Tables I and II show a higher average abundance of heavy water than do the results of EUCKEN and SCHÄFER. This can be explained, however, by taking into consideration the very slow motion of the Swedish glaciers. Thus, even in the upper parts of a glacier the ice is a residue of a several times larger bulk. It is necessary to acknowledge, in this instance, that even a sample from the névé of one of the glaciers had a density indicating an excess of heavy water (sample XIV, Table III). This sole sample does not constitute a certainty but possibly an enrichment is taking place already during the complicated process of snow recrystallizing into névé.

If the abundance of heavy water increases in the ice during the process of melting, it seems reasonable to suppose that the water in glacier streams ought to show a corresponding deficit as also follows from the thermodynamical calculations of EUCKEN and SCHÄFER. However, the density of some water running down the ice wall of the Kebnepakte glacier was found to be of quite a normal density (Table III). This is also the case with the water of lake Tarfala at the foot of the same glacier.

It ought also to be noted that one sample, taken from a small brook consisting of water from some large fields of melting snow, was of a density as high as some of the samples from glacier ice. This is, however, an isolated result which lacks sufficient corroboration. The object of the investigation presented here was only to ascertain whether there is an excess of heavy water in the ice of our Northern glaciers and whether it was possible, with comparatively simple means, to determine this excess. The collected material is too small to constitute the foundations for a thorough comparison with the results of EUCKEN and SCHÄFER or a discussion as to why some results are at variance with theory.

As a fact, it has been established that the ice of the Swedish glaciers carries a surplus of heavy water. This surplus can be as high as 0.05 per cent of the total bulk, a value corresponding to more than three times the normal abundance. As regards the measuring technique it seems possible to reach a higher degree of precision than is attainable with the present equipment.

The water samples were collected during summer tours in the mountains and I want to thank all those of my companions who aided me in collecting them. Also I thank Mr HARALD LILIEKVIST who, in his spare time, helped me with the measurements.

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REFERENCES. (1) **A. Eucken** and **K. Schäfer,** Göttingen Nachrichten, Mathemat.-Physikal. Klasse, Fachgruppe III, 1. Bd. p. 110, 1934; ibid. p. 138. — (2) **H. Moser,** Physik. Zeitschr. *35*, p. 682, 1934. — (3) **K. Linderstrøm-Lang, O. Jacobsen** and **G. Johansen,** C. R. Lab. Carlsberg *23*, p. 17, 1941. — (4) **A. E. Sandström** and **M. v. Gegerfelt,** Ark. matemat., astr. och fysik, *32 B*, No. 6, 1945.

Tryckt den 24 oktober 1951

Uppsala 1951. Almqvist & Wiksells Boktryckeri AB